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Kinetic model and Simulation Analysis for Propane Dehydrogenation in an Industrial Moving Bed Reactor

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***Abstract*—**A kinetic model for propane dehydrogenation in an industrial moving bed reactor is developed based on the reported reaction scheme. The kinetic parameters and activity constant are fine tuned with several sets of balanced plant data. Plant data at different operating conditions is applied to validate the model and the results show a good agreement between the model predictions and plant observations in terms of the amount of main product, propylene produced. The simulation analysis of key variables such as inlet temperature of each reactor (T_{inrx}) and hydrogen to total hydrocarbon ratio (H2/THC) affecting process performance is performed to identify the operating condition to maximize the production of propylene. Within the range of operating conditions applied in the present studies, the operating condition to maximize the propylene production at the same weighted average inlet temperature (WAIT) is ΔT_{inrx1}= -2, ΔT_{inrx2}= +1, ΔT_{inrx3}= +1 , ΔT_{inrx4}= +2 and ΔH2/THC= -0.02. Under this condition, the surplus propylene produced is 7.07 tons/day as compared with base case.

***Keywords*—**kinetic model, dehydrogenation, simulation, modeling, propane

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I. Introduction

In order for a company to remain world-class competitive it will be necessary to run the plant with less operating cost and at the same time, increase productivity. To realize this level of performance, it is crucial to simulate and optimize the entire process and plant. This requires a new level of understanding which includes the microkinetic models of each catalytic step. If entire processes are understood at this level, it will be possible to increase the output of most of our reactor systems between 50 and 100% and even up to 200 to 300% sometimes.

Dehydrogenation is a highly endothermic, equilibrium-controlled reaction. Equilibrium conversion and reaction rate increases with temperature, they are likewise favored at lower pressures because the volume of products exceeds that of reactants.

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In order to achieve reasonable economic conversion per pass (separation costs of unreacted paraffin are high), temperatures exceeding 550 °C are a prerequisite. High reaction temperature means magnification of side reactions. The stability of paraffins and olefins becomes critically influenced by the several side reactions. Oligomerization to heavier compounds, cracking to lighter hydrocarbons, skeletal isomerization, aromatization, alkylation of the formed aromatic rings, eventually leading to coke formation, lower the yields. Removal of hydrogen from the products improves the equilibrium extent and rate of dehydrogenation. However, recycle of hydrogen helps reduce the coke formation on the catalyst [1].

In view of the reaction characteristics as stated above, the optimum operating condition of the dehydrogenation reactor represents a compromise among the critical factors. In view of this, an accurate model and simulation tool is crucial in identifying the optimum operating condition of the plant [2].

In the present study, several type of kinetic and reactor models was validated using the inter reactor sample data. The best model was identified and rigorous simulations were performed to determine the operating condition for maximizing the production of propylene from dehydrogenation of propane.

II. Procedure

A. Type of Reactor Model Used

In the present study, all the chemical reactions possibly occurred in the reactor were incorporated into the reactor model for simulation. However, isomerization of iso-butane and dehydrogenation of iso-butane and n-butane were ignored due to its' negligible amount in the exit composition and unpredictable trend. Due to the lack of information, the rate expression to describe coke formation was not included in the present study. The amount of coke formed was assumed to be constant as long as the weighted average inlet temperature remained unchanged.

Since the slow moving bed reactors are employed, the reactors were modeled using plug flow reactor (PFR). In PFR or tubular flow reactor the feed enters at one end of a cylindrical tube and the product exits at the other end. The PFR model used in in-house software assumes there is no mixing in the axial direction and complete mixing in the radial direction. The PFR model is governed by the mole balances and design equations, rate law, stoichiometry, pressure drop

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correlations and energy balances equations. The flowsheet in Figure 1 shows the important steps of performing simulation using PFR in the in-house software.

B. Reaction Kinetics

The reaction schemes used in the present study is mainly based on the one developed by Loc et al. [3-4] and Lobera et al. [5-6] in addition to the side reactions proposed based on the inter reactor composition. The kinetic scheme for the propane reactions over the Platinum on Alumina catalyst are parallel network as below:

i. Main reaction (dehydrogenation reaction)

$C_3H_8 \leftrightarrow C_3H_6 + H_2$

(C3) (=C3) (H2)

ii. Side reaction (cracking reaction)

a. $C_3H_8 + H_2 \rightarrow C_2H_6 + CH_4$
(C3) (H2) (C2) (C1)

b. $C_2H_4 + H_2 \rightarrow C_2H_6$
(=C2) (H2) (C2)

c. $2C_3H_6 + H_2 \rightarrow C_6H_{14}$
(=C3) (H2) (C6)

d. $C_3H_6 + iC_4H_{10} \rightarrow CH_3C_6H_5 + 4H_2$
(=C3) (iC4) (T) (H2)

e. $C_3H_8 \rightarrow C_2H_4 + CH_4$

f. $C_3H_6 + H_2 \rightarrow C_2H_4 + CH_4$

III. KINETIC PARAMETERS ESTIMATION

A. Rate law developed by Loc et al. [3-4]

The equation describing the main reaction for propane dehydrogenation (reaction I) is taken from Loc et al. [3-4] and it is shown in (1):

$$r_I = a \times k_I \left(1 - \frac{P_{=C3} P_{H2}}{P_{C3} K_{eqI}}\right) \frac{P_{C3}}{P_{H2}^{0.5} + K_{=C3I} P_{=C3}} \text{ kmol/(s.m}^3\text{)} \quad (1)$$

Where K_{eqI} is the equilibrium constant of reaction I

$$K_{eqI} = 8.49e+8 \exp(-118707/(RT)) \text{ kPa} \quad (2)$$

and k_I and $K_{=C3I}$ is

$$k_I = 0.3874 \exp(-2950/T) \text{ kmol/(s.m}^3\text{.kPa}^{0.5}\text{)} \quad (3)$$

$$K_{=C3I} = 3.4785 \times 10^{-8} \exp(17200/T) (\text{kPa})^{-0.5} \quad (4)$$

Due to the lacking of information on the physical properties of catalyst, the dimensionless catalyst activity, *a* is fine tuned using the composition of the reactor inter stage sample. *a* for the 1st reactors is 0.34 while for the 2nd, 3rd and 4th reactors is 0.38.

Based on the literature, most of the rate law describing side reactions can be expressed in power law [3-6]. All the activation energy side reactions was taken from the literature while the pre-exponential factor of the Arrhenius equation was fine tuned base on the composition of the inter stage samples. Table I shows the rate laws for all the possible side reactions after fine tuning.

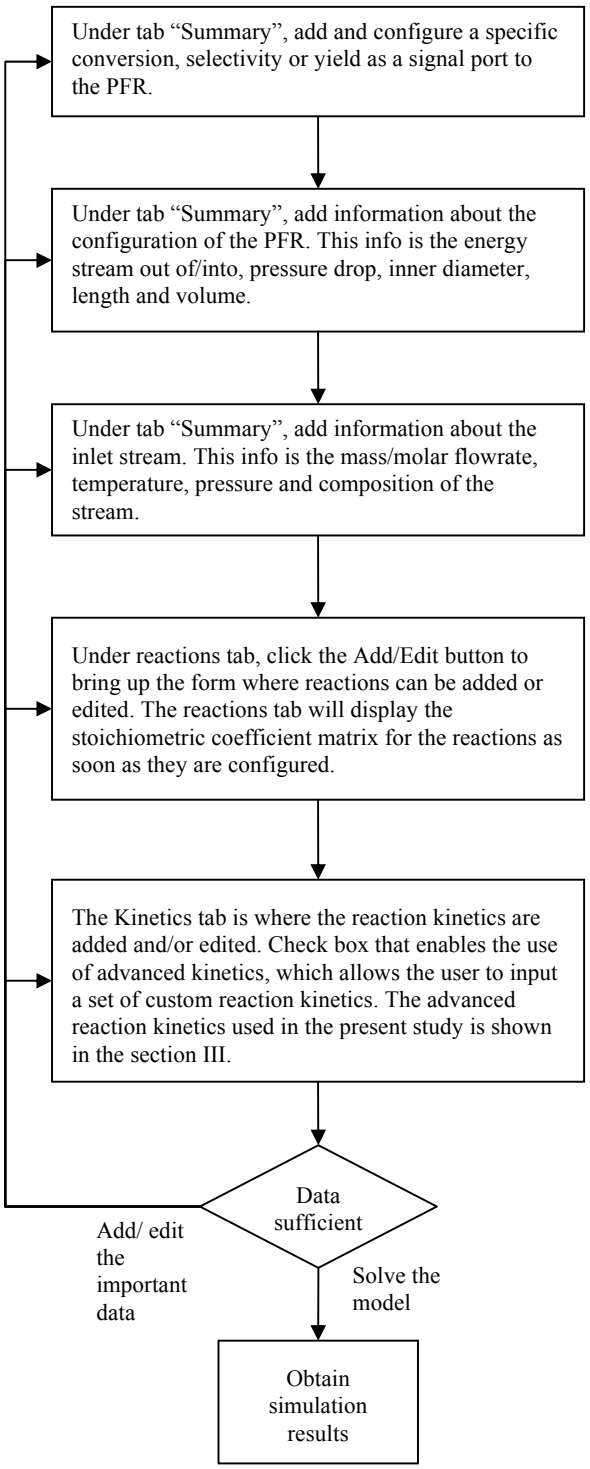


Fig. 1 Steps involved in the PFR simulations.

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TABLE I
THE FINE TUNED REACTION KINETICS OF THE SIDE REACTIONS USED SIMULTANEOUSLY WITH MAIN REACTION KINETICS DEVELOPED BY LOC ET AL. [3-4]

Side reaction	Rate law	Kinetic parameter	Reactor involved
a.	$r_{II} = k_{II} P_{C3} P_{H2}$	$k_{II} = 2.4e-9 \exp\left(\frac{-137000}{R}\left(\frac{1}{T}-\frac{1}{793.15}\right)\right)$	1-4
b.	$r_{III} = k_{III} P_{=C2} P_{H2}$	$k_{III} = 1.5E-7 \exp\left(\frac{-155000}{R}\left(\frac{1}{T}-\frac{1}{793.15}\right)\right)$	1-4
c.	$r_{IV} = k_{IV} P_{=C3} P_{H2}$	$k_{IV} = 3.0E-8 \exp\left(\frac{-18170}{RT}\right)$	2-4
d.	$r_{V} = k_{V} P_{=C3} P_{iC4}$	$k_{V} = 0.085 \exp\left(\frac{-50242}{RT}\right)$	2-4
e.	$r_{VI} = k_{VI} P_{C3}$	$k_{VI} = 33 \exp\left(\frac{-137000}{RT}\right)$	3-4
f.	$r_{VII} = k_{vii} P_{=C3} P_{H2}$	$k_{VII} = 1.55E+7 \exp\left(\frac{-256000}{RT}\right)$	3-4

B. Rate law developed by Lobera et al. [5-6]

The equation describing the main reaction for propane dehydrogenation (reaction 1) is taken from Lobera et al. [4-5] and it is shown in (5):

$$r_1 = a\frac{k_1\left(P_{C3} - (P_{=C3}P_{H2}/K_{eq})\right)}{1+(P_{=C3}/K_{=C3})} \quad (5)$$

Where K_{eq} is the equilibrium constant of reaction 1

$$K_{eq} = 8.49e+8\exp(-118707/(RT))\ \text{kPa} \quad (6)$$

and k_1 and $K_{=C31}$ is

$$k_1 = 5.15e-5\exp\left(\frac{-34570}{R}\left(\frac{1}{T}-\frac{1}{793.15}\right)\right)(\text{kmol/s.m}^3\text{.kPa}) \quad (7)$$

$$K_{=C31} = 349\exp\left(\frac{85817}{R}\left(\frac{1}{T}-\frac{1}{793.15}\right)\right)\ \text{kPa} \quad (8)$$

Due to the lacking of information on the physical properties of catalyst, the dimensionless catalyst activity, *a* is fine tuned using the composition of the reactor interstage sample. The *a* for the 1st, 2nd, 3rd and 4th reactors are 0.33, 0.47, 0.55 and 0.68 correspondingly.

Similar with the previous section, the reaction kinetics describing side reactions were fine tuned and it is shown in Table II.

In order to distinguish the PFR model incorporated with 2 different reaction kinetics, PFR model incorporated with reaction kinetics developed by Loc et al. [3-4] is labeled as PFR_Loc Model, whereas the PFR model incorporated with reaction kinetics developed by Lobera et al. [5-6] is labeled as PFR_Lobera Model.

TABLE II
THE FINE TUNED REACTION KINETICS OF THE SIDE REACTIONS USED SIMULTANEOUSLY WITH THE MAIN REACTION KINETICS DEVELOPED BY LOBERA ET AL. [4-5]

Side reaction	Rate law	Kinetic parameter	Reactor involved
a.	$r_2 = k_2 P_{C3} P_{H2}$	$k_2 = 2.2e-9\exp\left(\frac{-137000}{R}\left(\frac{1}{T}-\frac{1}{793.15}\right)\right)$	1-4
b.	$r_3 = k_3 P_{=C2} P_{H2}$	$k_3 = 1.2E-7\exp\left(\frac{-155000}{R}\left(\frac{1}{T}-\frac{1}{793.15}\right)\right)$	1-4
c.	$r_4 = k_4 P_{=C3} P_{H2}$	$k_4 = 2.8E-8\exp\left(\frac{-18170}{RT}\right)$	2-4
d.	$r_5 = k_5 P_{=C3} P_{iC4}$	$k_5 = 0.085\exp\left(\frac{-50242}{RT}\right)$	2-4
e.	$r_6 = k_6 P_{C3}$	$k_6 = 29\exp\left(\frac{-137000}{RT}\right)$	3-4
f.	$r_7 = k_7 P_{=C3} P_{H2}$	$k_7 = 1.35E+7\exp\left(\frac{-256000}{RT}\right)$	3-4

IV. Results and Discussion

A. Base Case Simulations

The reactor system in the present study comprises of 4 adiabatic moving bed reactors in series with interstage re-heating in fired furnaces. For regeneration, the catalyst slowly downflows, and it is collected at the end of the last reactor, conveyed to the regenerator (CCR) and then transferred back to the first reactor. The reactor temperature profile is a typical sequence of reheating steps [7-9]. Table III shows the operating conditions of each reactor for the base case. The simulation results are shown in Figure 3, Figure 4 and Table IV.

TABLE III
OPERATING CONDITION OF THE BASE CASE (PLANT DATA ON 18 NOV 2010)

Reactor No.	Rx 1	Rx 2	Rx 3	Rx 4
Inlet temperature (°C)	*A	B	C	D
Inlet pressure (kPa)	E	F	G	H
H2/THC ratio (mol/mol)	J			

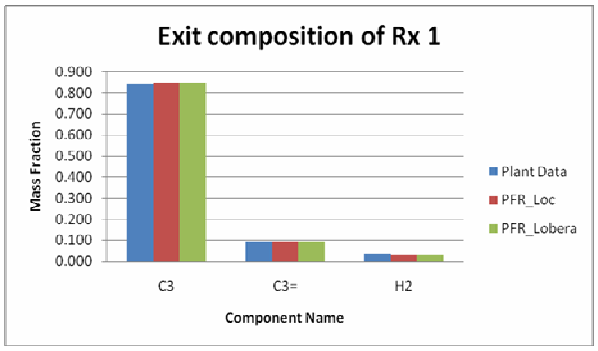
*Due to its' confidentiality, all the operating condition is given as unknown with the range of: 630<A<D<B<C; 150<H<G<F<E, J<0.60

As can be seen from Figure 3 and 4, all the models used gives comparable predictions to the exit compositions, as indicated by the comparable absolute relative error in Table IV.

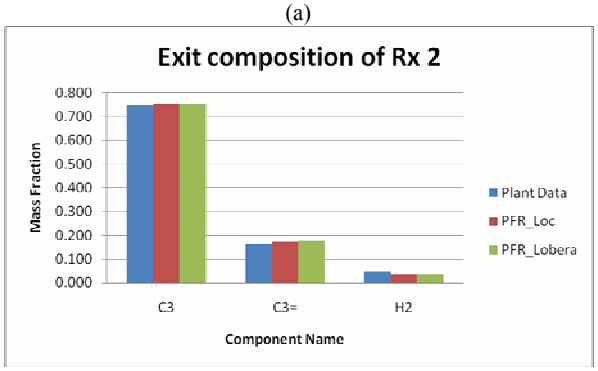
Between the models studied, PFR_Loc model gives better prediction to the exit composition of all the reactants and products with the average absolute relative error (AARE) of 9.83%, as shown in Table IV. Despite the acceptable range of AARE, the deviations of the predicted composition of H2, =C2 and C2 from the plant data are 21%, 14% and 11% respectively. These significant deviations are due to the assumptions of no radial variations in velocity, concentration and temperature or reaction rate in the reactor during the

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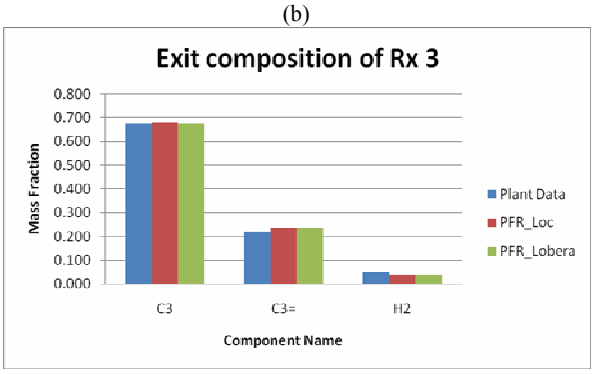
simulation. The simulation was done solely based on the axial variations.



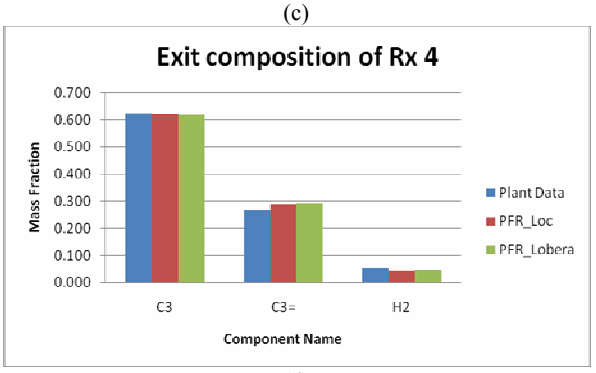
(a)



(b)

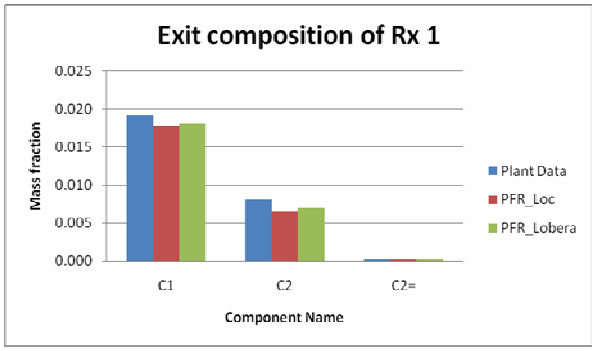


(c)

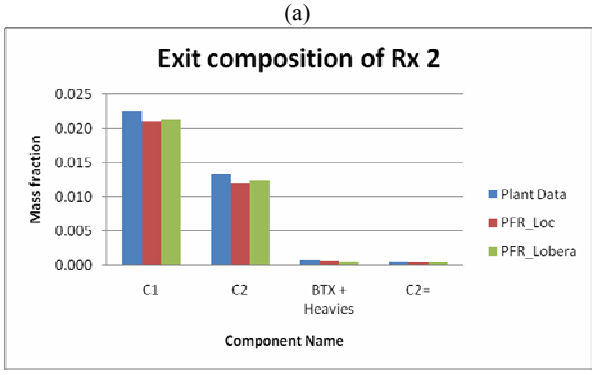


(d)

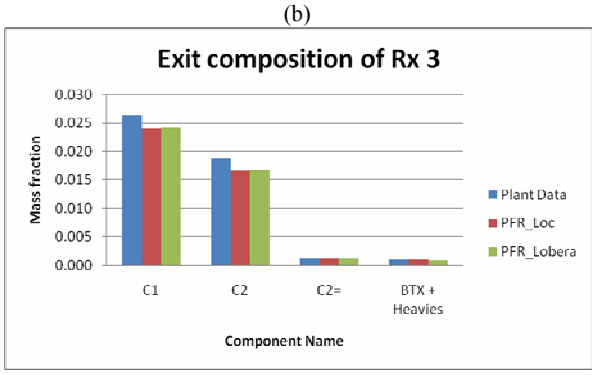
Fig. 3 Product and reactant compositions of the main reaction in the reactor exit stream



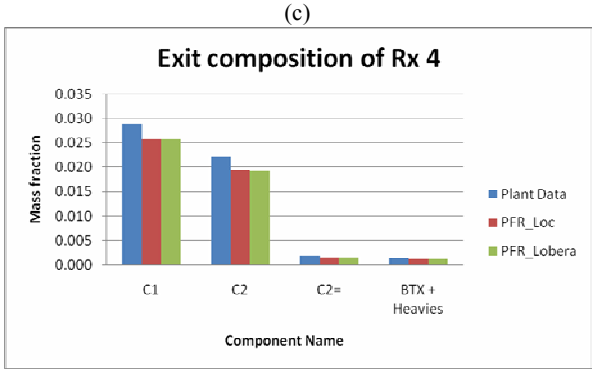
(a)



(b)



(c)



(d)

Fig. 4 Product compositions of the side reactions in the reactor exit stream

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TABLE IV AARE FOR THE MODELS USED IN EXIT COMPOSITION PREDICTION.

Component	Base Case	Case 1
PFR_Loc		
C3	0.69%	0.65%
C3=	5.39%	4.18%
H2	20.63%	7.14%
C1	8.20%	7.83%
C2	13.62%	5.97%
C2=	11.48%	11.97%
BTX + Heavies	8.77%	18.43%
AARE	9.83%	8.02%
PFR_Lobera		
C3	0.65%	0.42%
C3=	6.07%	4.09%
H2	20.54%	7.31%
C1	7.56%	7.50%
C2	11.08%	8.40%
C2=	11.94%	13.17%
BTX + Heavies	12.35%	22.16%
AARE	10.03%	9.01%

Figure 5 shows the yield per pass profiles of the entire reactor system. The increase in temperature from reactor 1 to 4 has increased the conversion as more propane is converted to the main and side products. Nevertheless, the selectivity from reactor 1 to 4 is suppressed by the increase of temperature because high temperature favors the formation of side products. As a whole, the yield per pass was increasing throughout the entire reactor system, as can be seen from Figure 5. Since PFR_Loc model could predict the composition of propane and propylene with smaller absolute relative error of <5.5% as shown in Table IV, the yield per pass is also better described by this model.

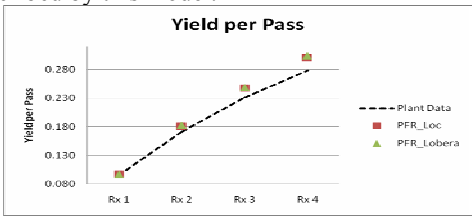


Fig. 5 Yield per pass profile throughout the entire reactor system.

B. Model Validation with Different Cases

In order to test the consistency of the models, the operating conditions were varied. For Case 1, the inlet temperature of the Reactor 1 was decreased 1 deg. C while the inlet temperature of Reactor 4 was increased 1 deg. C. The operating condition and composition of the product were the average value of 5 days (25 Nov 2010, 6 Dec 2010, 10 Dec 2010, 13 Dec 2010 and 16 Dec 2010). The same models were adopted for the simulations and AARE for all the models in exit composition prediction of both cases is compared in Table IV. It is found that all the models could predict the composition of case 1 with lower AARE and PFR_Loc Model offers the least AARE. In addition to the composition validation under different cases, the best model, PFR_Loc Model also was tested using the archive data in terms of its consistency in predicting the amount of propylene produced. Table V shows the operating conditions of all the cases selected for the study.

TABLE V
OPERATING CONDITIONS OF ALL THE SELECTED CASES FOR THE VALIDATION OF THE AMOUNT OF PROPYLENE PRODUCED.

Case No.	Inlet temperature difference, ΔT_{inrx}				H2/THC molar ratio difference, ΔH2/THC
	Rx 1	Rx 2	Rx 3	Rx 4	
2 (22 Dec 10)	*-1.33	-0.05	-0.05	1.42	0.00
3 (9 July 10)	-0.07	-0.21	-2.95	0.03	0.00
4 (20 Apr 10)	0.24	0.22	0.36	0.44	0.04
5 (25 Apr 10)	-0.02	-0.16	-0.09	0.06	0.05
6 (26 Apr 10)	0.11	-0.16	-0.10	0.42	0.06

*Due to its' confidentiality, all the number in the table is given as the difference with base case.

From Table VI, the deviation of the predicted amount of propylene produced from plant data is approximately 4.41% (absolute relative error, ARE). The results also show that the changes predicted by the model is identical to the changes generated from the plant data. These changes are referring to the changes of propylene flow rate in liquid product as compared with the base case. In case 2, more propylene has been produced with a decrease in inlet temperature of the 1st reactor and an increase in the inlet temperature of the 4th reactor. In case 3, a reduction in the inlet temperature of the 3rd reactor has caused a reduction in the propylene production. From case 4 to case 6, the increase in the H2/THC ratio has reduced the amount of propylene in the liquid product. All these changes are observed due to the nature of the dehydrogenation reaction, which is endothermic equilibrium limited. Higher temperature and lower H2/THC ratio shift the reaction to the forward direction. Hence, more propylene is produced.

C. Simulation Analysis

1. Effect of reactor inlet temperature

The inlet temperature of each reactor in the entire reactor system was varied by ±2 °C while the H2/THC ratio, inlet flow rate and inlet stream composition were kept constant. The simulation results show that reactor system with higher WAIT will produce more propylene and vice versa if it is operated at lower WAIT as compared with base case. Higher temperature favors the forward reaction of the highly endothermic dehydrogenation reaction and hence more

propylene will be produced. Part of the simulation results under the same WAIT have been sorted out and tabulated in Table VII.

TABLE VI
COMPARISON OF THE AMOUNT OF PROPYLENE PRODUCED BETWEEN PLANT DATA AND RESULTS PREDICTED BY THE MODEL SIMULATION

Case No.	Propylene in liquid product, kg/hr	Propylene in liquid product for base case, kg/hr	Changes in propylene flow rate as compared with base case, kg/hr	
	ARE, %	ARE, %	plant data	Model
2	3.99	4.15	*+	66.79
3	4.54	4.15	-	-557.02
4	4.49	4.15	-	-169.05
5	4.34	4.15	-	-289.52
6	4.70	4.15	-	-199.23

*+ represents surplus, - represents shortage

Base on the data no. 4-6, 9-10, 12, 16-18, 21-22 and 24 in Table VII, the amount of propylene reduced due to the decrease of inlet temperature of one reactor can be compensated by the amount of propylene produced by the following reactor with the increase of temperature. As compared with the base case, surplus propylene can be produced under these operating conditions because of the difference in the degree of sensitivity of equilibrium conversion to reactor temperature (S). Based on the data given by Cavani and Trifiro [10], the equilibrium conversion is most sensitive to the temperature when the temperature is ranged at 600-650 °C. Therefore, S of the reactor system is ranked as $S_{4th\ reactor}>S_{3rd\ reactor}>S_{2nd\ reactor}>S_{1st\ reactor}$. An increase of 1 °C in the inlet temperature of 4th reactor could produce more propylene if comparing with an increase of 1 °C in the inlet temperature of 1st, 2nd and 3rd reactors.

Under the identical WAIT, the maximum amount of propylene could be produced when the inlet temperature differences from the base case for 1st, 2nd, 3rd and 4th reactor are -2 °C, +1 °C, +1 °C and +2 °C respectively. The amount of propylene produced is an additional of 6.5 tons/day as compared to the base case. The reduction of the inlet temperature of the 1st reactor has reduced the amount of main and side products. Nevertheless, the amount of propylene and side products produced by the reactor system is more as compared with base case because the reduction in the 1st reactor has been compensated by the increment in the 2nd, 3rd and 4th reactors with the increase of inlet temperature.

2. Effect of H2/THC Ratio

The H2/THC ratio of the entire reactor system was varied by ±0.02 while the inlet temperature, inlet flow rate and inlet stream composition were kept constant. From Table VIII, the simulation results show that reactor system with lower H2/THC ratio will produce more propylene and vice versa if it is operated at higher H2/THC ratio as compared with base case. The reactor system operates at H2/THC ratio difference of -0.02 (data no. 5 in Table VIII) gives maximum surplus of propylene production as compared with the base case, which is 0.56 tons/day. Lower H2/THC ratio shifts the reaction equilibrium to the product side and hence more propylene will be produced.

TABLE VII
SIMULATION RESULTS FOR THE STUDY OF EFFECT OF INLET TEMPERATURE

No.	ΔT_{inrx}				Difference in mass flow rate as compared with base case, tons/ day
	Rx 1	Rx 2	Rx 3	Rx 4	
1	*1	-1	0	0	-0.65
2	1	0	-1	0	-0.97
3	1	0	0	-1	-1.15
4	-1	1	0	0	0.66
5	-1	0	1	0	0.99
6	-1	0	0	1	3.40
7	0	1	-1	0	-0.31
8	0	1	0	-1	-0.49
9	0	-1	1	0	0.33
10	0	-1	0	1	0.51
11	0	0	1	-1	-0.16
12	0	0	-1	1	0.20
13	2	-2	0	0	-1.29
14	2	0	-2	0	-1.91
15	2	0	0	-2	-2.27
16	-2	2	0	0	1.34
17	-2	0	2	0	1.99
18	-2	0	0	2	2.36
19	0	2	-2	0	-0.60
20	0	2	0	-2	-0.95
21	0	-2	2	0	0.68
22	0	-2	0	2	1.06
23	0	0	2	-2	-0.30
24	0	0	-2	2	0.42

**Due to its' confidentiality, all the inlet temperature in the table is given as the difference with base case.

TABLE VIII
SIMULATION RESULTS OF THE STUDY OF EFFECT OF H2/THC RATIO

No.	ΔH2/THC	Difference in mass flow rate as compared with base case, tons/ day
1.	*0.02	-0.58
2.	0.01	-0.29
3.	0.00	0.00
4.	-0.01	0.29
5.	-0.02	0.56

**Due to its' confidentiality, all the H2/THC in the table is given as the difference with base case.

3. Effect of the combination of reactor inlet temperature and H2/THC ratio

The H2/THC ratio and inlet temperature of the entire dehydrogenation system were varied by ±2 °C and ±0.02 respectively, while the inlet flow rate and inlet stream composition were kept constant. The simulation results show that the operating condition that maximize the production of propylene is ΔT_{inrx1}= -2, ΔT_{inrx2}= +1, ΔT_{inrx3}= +1, ΔT_{inrx4}= +2

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and ΔH2/THC= -0.02. Under this condition, the surplus propylene produced is 7.07 tons/day.

V. Conclusion

Among the models studied, model PFR_Loc offers the least AARE for the composition prediction of the dehydrogenation system under various operating conditions. Model PFR_Loc is robust to predict the changes in composition when there are changes in the operating conditions.

Higher temperature and lower H2/THC shifts the dehydrogenation reaction to the product side and hence more propylene will be produced.

In the reactor system at the same WAIT, it is preferably to increase the temperature of the reactors which operate at higher range of temperature to obtain more surplus propylene as compared with the base case (e.g., increase the temperature of reactor 2 could have more surplus propylene as compared with increasing the temperature of reactor 1; increase the temperature of reactor 3 could have more surplus propylene as compared with increasing the temperature of reactor 2).

Within the range of operating conditions applied in the present study, the operating condition to maximize the propylene production is ΔT_{inrx1}= -1, ΔT_{inrx2}= +1, ΔT_{inrx3}= +1, ΔT_{inrx4}= +2 and ΔH2/THC= -0.02. Under this condition, the surplus propylene produced is 7.07 tons/day as compared with base case.

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